Title	Electrically Tunable Mirrorless Laser Based on a Dye-Doped Ferroelectric Liquid Crystal
Author(s)	Ozaki, Masanori; Kasano, Masahiro; Yoshino, Katsumi et al.
Citation	電気材料技術雑誌. 11(1) p.67-p.72
Issue Date	2002-05-25
oaire:version	VoR
URL	https://hdl.handle.net/11094/81700
rights	
Note	

Electrically Tunable Mirrorless Laser Based on a Dye-Doped Ferroelectric Liquid Crystal

Masanori OZAKI, Masahiro KASANO and Katsumi YOSHINO

Department of Electronic Engineering, Graduate School of Engineering,
Osaka University, 2-1 Yamada-Oka, Suita, Osaka 565-0871 Japan

Dirk GANZKE and Wolfgang HAASE

Institute of Physical Chemistry, Darmstadt University of Technology
Petersenstr. 20, Darmstadt 64287 Germany

Optically pumped laser emission has been observed at the edge of one-dimensional photonic band of dye-doped chiral smectic liquid crystal with a periodic helical structure which is a so-called ferroelectric liquid crystal. Lasing wavelength was successfully tuned upon applying electric field in the direction perpendicular to the helix axis, which is based on the change in helix periodicity caused by the deformation of the helix upon the field application.

KEYWORDS : ferroelectric liquid crystal, photonic crystal, laser, stop band

電界により発振波長制御可能な色素ドープ強誘電性液晶レーザー

尾崎　雅則、笠野　真弘、吉野　勝美

大阪大学大学院工学研究科電子工学専攻
〒565-0871 大阪府吹田市山田丘2-1

ダーク・ガンツケ、ヲルフガング・ハーゼ

Institute of Physical Chemistry, Darmstadt University of Technology
Petersenstr. 20, Darmstadt 64287 Germany

色素をドープしたカイラルスメクチック液晶（強誘電性液晶）を光励起したところ、螺旋周期構造に起因するストップバンドの端からレーザー発振を観測した。螺旋軸に垂直方向に電界を印加して螺旋ピッチの変化すなわちストップバンドをシフトさせることにより、レーザー発振波長を広範囲に変化させることに成功した。

1.introduction

Photonic crystals having an ordered structure with a periodicity of optical wavelength have attracted considerable attention from both fundamental and practical points of view, because novel physical concepts such as photonic band gap have been theoretically predicted and various applications of photonic crystals have been proposed.[1-2] Especially, the study of stimulated emission in photonic band gap is one of the most attractive subjects, since, in the band gap, a spontaneous emission is inhibited and low-threshold lasers based on photonic crystals are expected.[2-7] In a one-dimensional periodic structure, the laser action has been expected at the photonic band edge where the photon group velocity approaches zero.[8] In order to realize the photonic crystal, a large number of intensive studies on a micro-fabrication based on a semiconductor processing technology and a self assembly construction of nano-scale spheres have been carried out.

Liquid crystals including chiral molecule have a self-organized helical structure which is a one-dimensional periodic structure and shows characteristic optical properties.[9] In cholesteric liquid crystals with helical structure, light propagating along the helical axis is selectively reflected depending on the polarization states if the wavelength of the light matches to the optical periodicity of the helical structure, which is a so-called selective reflection. The wavelength region in which the light can not propagate is the stop band, which is considered as a one-dimensional pseudo-bandgap. Lasing at the band edge has been reported in the cholesteric liquid crystal.[10-12]

Chiral smectic liquid crystals with tilted structure show a ferroelectricity, which is called ferroelectric liquid crystal (FLC), and have an expected potential for the electrooptic applications because of the fast response to the electric field.[13] The FLC also has a helical structure and shows the selective reflection due to the one-dimensional periodic structure as the almost same manner as the cholesteric liquid crystal,[14] and the laser action in the helical structure of FLC has been demonstrated.[15] The helix of FLC can be easily deformed upon applying electric field and its response is fast because of the strong interaction between the spontaneous polarization and electric field. In this study, we prepare FLC mixtures having a well-controlled short pitch of the helical structure whose periodicity is equivalent to the visible optical wavelength, and successfully demonstrate a lasing wavelength tuning in a wide range upon applying electric field for the first time.

2. Experiment

The FLC compound used in this study is a multi-component mixture having the chiral smectic C (SmC*) phase in a wide temperature range including a room temperature (~0℃ to 68℃). A molecular tilt angle and spontaneous polarization at 30℃ are 26° and 55nC/cm^2, respectively. As a laser dye doped in the FLC, a Coumarin 500 (Exciton) was used. The concentration of the dye is 0.2 wt%. The sample was filled into a sandwich cell which consists of two glass plates. Aluminum foils were used as both spacers determining cell thickness and electrodes, which allows us to apply the electric field parallel to the glass substrates. The cell gap was 50μm. The electrodes distance is 2mm. In order to obtain a homeotropically aligned cell, surfaces were coated with a polyimide (JALS-2021-R2, Japan

Synthetic Rubber). In the homeotropically aligned cell, the helicoidal axis is perpendicular to the glass substrates, and the electric field is applied parallel to the smectic layer and perpendicular to the helix axis. The temperature of the sample cell was controlled using a hot bath and temperature controller. The transmission spectrum measurement was performed using a spectrometer (Shimadzu, UV-3100PC).

For an excitation source of emission measurement, second harmonic light of a regenerative amplifier system based on a Ti:sapphire laser (Spectra Physics) was used. The pulse width, wavelength and pulse repetition frequency of the output laser beam were 150fs, 400nm and 1 kHz, respectively. The excitation energy can be varied within the range from 0.01 to 28 μJ/pulse. The illumination area on the sample was about 0.2 mm^2. The excitation laser beam irradiated the sample at an angle of 45° with respect to the cell plate normal. The emission spectra from the dye-doped FLC were measured from the opposite side of the cell using a spectrograph (Oriel MS257) with a CCD detector having spectral resolution of 0.3nm was used. The collecting direction was perpendicular to the cell surface, which is normal to the smectic layers and along the helical axis.

3. Results and Discussion

When the energy of excitation laser beam is low ($\leq$ 1.76 μJ/pulse), the emission spectrum is dominated by a broad spontaneous emission and the dips are observed in the broad spectra. The wavelength of the dip coincides with that of the stop band for the half pitch of the helix of FLC. As the excitation energy increases, the emission intensity is enhanced. When the excitation energy is over the threshold, laser action is observed as a sharp peak at the edge of the dip. The full width at half maximum (FWHM) of the emission peak is about 0.5nm, which is limited by the spectral resolution of our experimental setup.

Although, at a low excitation energy, only a scattered flash of the was observed, a clear emission spot appears at high excitation, which indicates a sharp directional emission characteristics from the FLC laser. It should be noted that no mirrors are needed for the directional laser emission, indicating that a mirrorless lasing is achieved. In addition, for the lasing, the film is not needed to be fixed on a stable mount and to be well-aligned.

The emission energy dependences of peak intensity and linewidth of the emission spectrum have also been studied, and the presence of a lasing threshold was confirmed. At lower excitation energy, the emission intensity increases in proportion to the pump energy. Above the threshold at a pump pulse energy of about 3mJ, the emission intensity non-linearly increases. The linewidth of the emission spectrum also drastically decreases above the threshold. These results confirm that lasing occurs above the threshold of the pump energy at the edge of the

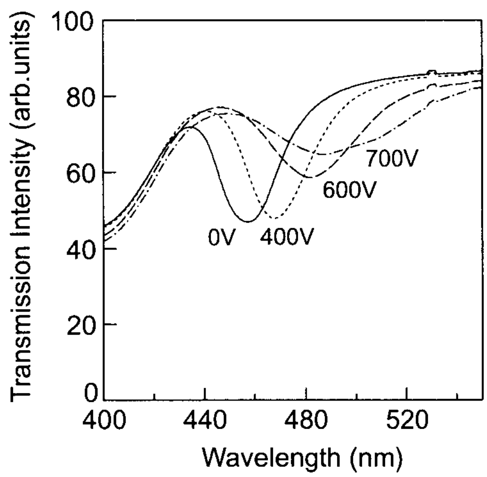


Fig.1 Transmission spectra of a homeotropically aligned FLC cell as a function of the voltage applied parallel to the glass substrates of the sample cell.

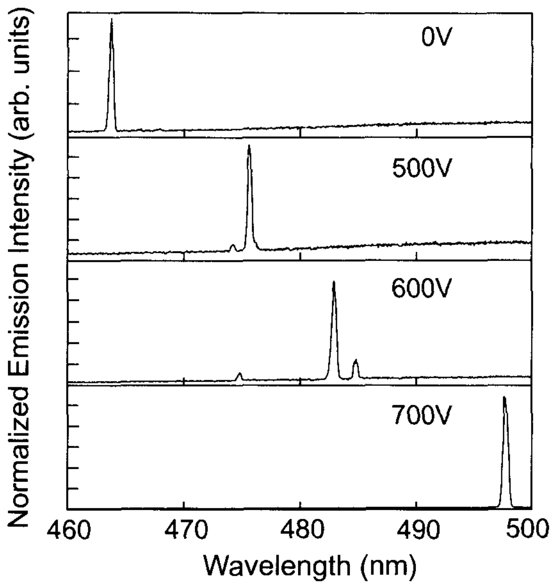


Fig.2 Lasing spectra of a dye-doped FLC at high excitation energy (24μJ/pulse) as a function of the applied voltage.

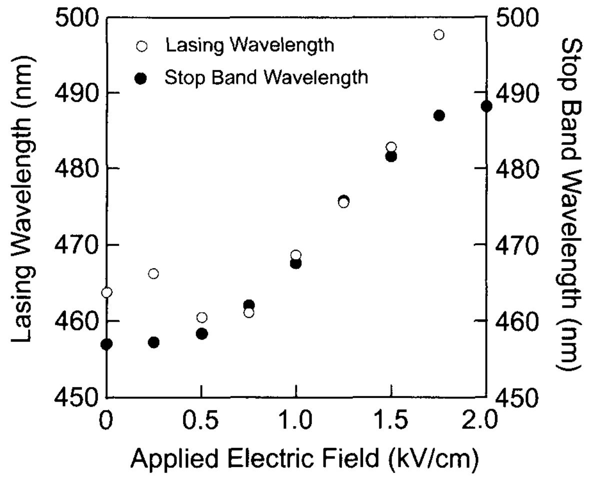


Fig.3 The electric field dependence of the lasing wavelength (open circles) and stop band wavelength (closed circles) of dye-doped FLC.

photonic stop band in the spontaneous emission. The threshold pump energy for lasing would be lowered by optimizing a doped laser dye, a concentration of the dye, a molecular tilt angle of FLC with respect to the smectic layer and cell geometries such as the thickness and smectic layer arrangement. The design of molecular structure of the host FLC for a high birefringence might be one of the most important factors for the low threshold lasing.

The periodicity of the helix can be controlled upon applying electric field perpendicular to the helical axis. FLC has a spontaneous polarization P_s which points normal to the molecules and parallel to the smectic layers. When the electric field is applied in the layer, P_s intends to point along the field direction and all FLC molecules orients to the same direction normal to the field, resulting in the deformation of the helix.

Figure 1 shows the transmission spectra of the homeotropically aligned FLC film as a function of the voltage applied parallel to the glass substrates of the sample cell. In this geometry, the electric field is applied perpendicular to the helical axis. As is evident from Fig. 1, the transmission dip due to the selective reflection shifts toward longer wavelength with increasing voltage. This means that the helix is elongated upon the field application.

The fact that the periodicity of the helical structure of the dye-doped FLC can be controlled upon the field application allows us to expect the electric field tuning of the laser emission wavelength. Figure 2 shows the lasing spectra of the dye-doped FLC at high excitation energy (24μJ/pulse) as a function of the applied voltage. It should be noted that lasing wavelength largely shifts toward longer wavelength with increasing voltage, which corresponds to the shift of the selective reflection band. In spite of a low field (3.5kV/cm), a wide tuning of the lasing wavelength was achieved.

Figure 3 shows field dependence of the lasing wavelength. The selective reflection wavelength is also shown as a function of the applied electric field. It can be confirmed that the shift of the lasing wavelength is based on the band shift caused by field-induced change in the helix periodicity.

The elctrooptic effects originating from the field-induced deformation of the helix in the FLC have been proposed.[16,17] Especially, the response of the electrooptic switching based on a small deformation of the helix of a short pitch FLC is as fast as several ms order and the application to an optical communication as well as to a display device has been proposed.[18] The relaxation times of the helix deformation of FLC is represented by the following equation,

$$\tau \propto \frac{p^2\gamma}{K},$$

where p is the helix pitch, γ is the rotational viscosity and K is the elastic constant.

According to this relation, the response time is inversely proportional to p^2, and high frequency modulation of the periodicity of the helix can be expected in a short pitch FLC. In deed, the electrooptic modulation device using a similar compound to the FLC material used in this study has of the order of several μs response. Consequently, the fast modulation of the lasing wavelength is possible in the dye-doped FLC with a short pitch. The applications to the light source of the optical communication and the Q-switched or active mode-locked lasers using this FLC can be expected.

4. Conclusions

In conclusions, optically pumped laser emission was observed without any mirrors at the edge of one-dimensional photonic band of dye-doped chiral smectic liquid crystal with a periodic spiral structure. Lasing wavelength was widely tuned by adjusting the periodicity of the helical structure upon applying electric field. The large shift of the lasing wavelength by 40nm was achieved by the application of a relatively low electric field.

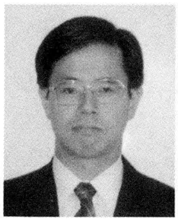
Acknowledgment

This work is supported by a Grant-in-Aid for Scientific Research from the Japan Ministry of Education, Culture, Sports, Science and Technology (14350165).

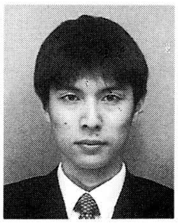
References

1) S. John: Phys. Rev. Lett., 58 (1987) 2486.
2) E. Yablonovitch: Phys. Rev. Lett., 58 (1987) 2059.
3) J.Martorell, N.M.Lawandy: Phys. Rev. Lett., 65 (1990) 1877.
4) K. Yoshino, K. Tada, M. Ozaki, A. A. Zakhidov, R. H. Baughman: Jpn. J. App. Phys., 36 (1997) L714.
5) K.Yoshino, S.B.Lee, S.Tatsuhara, Y.Kawagishi, M.Ozaki, A.A.Zakhidov: Appl. Phys. Lett., 73 (1998) 3506.
6) K.Yoshino, S.Tatsuhara, Y.Kawagishi, M.Ozaki, A.A.Zakhidov, Z.V.Vardeny: Appl. Phys. Lett., 74 (1999) 2590.
7) Y.A.Vlasov, K.Luterova, I.Pelant, B.Honerlage, V.N.Astratov: Appl.Phys.Lett., 71 (1997) 1616.
8) J.P.Dowling, M.Scalora, M.J.Bloemer, C.M.Bowden: J.Appl.Phys., 75 (1994) 1896.
9) S.Chandrasekhar: Liquid Crystals, Cambridge University Press, 1992.
10) V.I.Kopp, B.Fan, H.K.Vithana, A.Z.Genack: Opt.Lett., 23 (1998) 1707.
11) B.Taheri, A.F.Munoz, P.Palffy-Muhoray, R.Twieg: Mol.Cryst.Liq.Cryst., 358 (2001) 73.
12) H.Finkelmann, S.T.Kim, A.Munoz, P.Palffy-Muhoray, B.Taheri: Adv.Mater., 13 (2001) 1069.
13) J.W.Goodby, R.Blinc, N.A.Clark, S.T.Lagerwall, M.A.Osipov, S.A.Pikin, T.Sakurai, K.Yoshino, B.Zeks: Ferroelectric Liquid Crystals, Gordon and Breach Science Publishers, Philadelphia, 1991.
14) K.Hori: Mol.Cryst.Liq.Cryst., 82 (1982) L13.
15) M.Ozaki, M.Kasano, D.Ganzke, W.Haase and K.Yoshino: Adv.Mater., 14 (2002) 306.
16) K.Yoshino, K.G.Balakrishnan, T.Uemoto, Y.Iwasaki and Y.Inuishi: Jpn.J.Appl.Pjys., 17 (1978) 597.
17) L.A.Beresnev, V.G.Chigrinov, D.I.Dergachaev, E.P.Pozhidaev, J.Funfschilling and M.Schadt: Liq.Cryst., 5 (1989) 1171.
18) E.Pozhidaev, S.Pikin, D.Ganzke, S.Shevtchenko and W.Haase: Ferroelectrics, 246 (2000) 235.

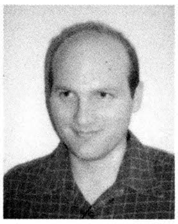
(Mar.1,2002 accepted)



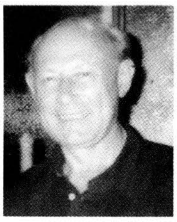
Masanori Ozaki was born in Aichi, Japan on February 26, 1960. He graduated in 1983 from Department of Electrical Engineering, Osaka University, where he obtained a Doctor of Engineering Degree in 1988. In that year, he became an Research Associate in Osaka University and promoted to Lecturer in 1991. In 1994, he became an Associate Professor, Osaka University.



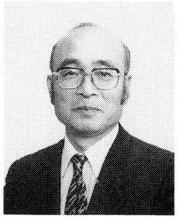
Masahiro Kasano was born in Wakayama, Japan on December 10, 1978. He graduated in 2001 from Department of ELectronic Engineering, Osaka University. He is now Master course student of Department of Electronic Engineering, Osaka University.



Dirk Ganzke was born in Berlin, Germany on September 9, 1972. He is now PhD student of Department of Physical Chemistry, Technical University of Darmstadt.



Wolfgang Haase was born in Born, Germany on October 25, 1936. He graduated in 1957 from Chemical Engineering, University of Magdeburg. He obtained a Doctor of Engineering Degree from University of Jena in 1964. In 1971, he became a Professor in Department of Physical Chemistry, Technical University of Darmstadt.



Katsumi Yoshino was born in Shimane, Japan on December 10, 1941. He graduated in 1964 from Department of Electrical Engineering, Faculty of Engineering, Osaka University, where he obtained a Doctor of Engineering Degree in 1969. In that year, he became an Assistant in Electrical Engineering, Osaka University. He promoted to Lecturer in 1972, Associate Professor in 1978. In 1988, he became a Professor in Department of Electronic Engineering, Faculty of Engineering, Osaka University.